Fukushima-derived radionuclides in the ocean and biota off Japan

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The Tōhoku earthquake and tsunami of March 11, 2011, resulted in unprecedented radioactivity releases from the Fukushima Dai-ichi nuclear power plants to the Northwest Pacific Ocean. Results are presented here from an international study of radionuclide contaminants in surface and subsurface waters, as well as in zooplankton and fish, off Japan in June 2011. A major finding is detection of Fukushima-derived ^{134}Cs and ^{137}Cs throughout waters 30–600 km offshore, with the highest activities associated with near-shore eddies and the Kuroshio Current acting as a southern boundary for transport. Fukushima-derived Cs isotopes were also detected in zooplankton and mesopelagic fish, and unique to this study we also find ^{110m}Ag in zooplankton. Vertical profiles are used to calculate a total inventory of ∼2 PBq ^{137}Cs in an ocean area of 150,000 km^2. Our results can only be understood in the context of our drifter data and an oceanographic model that shows rapid advection of contaminants further out in the Pacific. Importantly, our data are consistent with higher estimates of the magnitude of Fukushima fallout and direct releases [Stohl et al. (2011) *Atmos Chem Phys Discuss* 11:28319–28394; Bailly du Bois et al. (2011) *J Environ Radioact*, 10.1016/j.jenvrad.2011.11.015]. We address risks to public health and marine biota by showing that though Cs isotopes are elevated 10–1,000× over prior levels in waters off Japan, radiation risks due to these radionuclides are below those generally considered harmful to marine animals and human consumers, and even below those from naturally occurring radionuclides.

The loss of power and subsequent overheating, meltdowns, and hydrogen explosions at the Fukushima Dai-ichi nuclear power plants (NPPs) resulted in airborne fallout over land and the ocean that peaked in mid-March 2011 (1–3). In addition to atmospheric fallout, waters used to cool the reactors that subsequently leaked from NPP buildings provided a direct path for contamination of the oceans (4). Concentrations at the NPP ocean discharge channels peaked in early April at more than 50 million times preexisting ocean levels of ^{137}Cs (5). Though considerable data have been released in Japanese reports regarding the concentration of selected radionuclides in the air, soil, and coastal discharge sites, large uncertainties remain, including even the magnitude of total atmospheric releases (6) and direct discharges (7). There is also little information on radionuclide distributions offshore to help assess contamination and transport in the North Pacific and for independent confirmation of whether the levels are of human health concern.

We report here on a comprehensive analysis of radionuclide concentrations off Japan in surface and subsurface waters as well as for mesopelagic fish and plankton collected from the R/V *Ka'imikai-o-Kanaloa* at 50 stations June 4–18, 2011 (Fig. S1). We used standard oceanographic methods to collect water and biota followed by laboratory processing and detection of marine radionuclides using high-purity germanium spectroscopy (*Methods* and *SI Methods*). This report describes the presence of the gamma-emitting isotopes ^{134}Cs ($t_{1/2}$ = 2.07 y), ^{137}Cs ($t_{1/2}$ = 30.07 y), and ^{110m}Ag ($t_{1/2}$ = 250 d). With their shorter half-lives, any ^{134}Cs and ^{110m}Ag seen in our samples could only be derived from the 2011 Fukushima NPP releases.

Cesium is a highly seawater soluble radionuclide whose primary source to the ocean before March 2011 has been from weapons testing in the 1960s, with lesser amounts from Chernobyl fallout in 1986 and intentional discharges such as from European nuclear fuel reprocessing facilities at Cap de la Hague (France) and Sellafield (United Kingdom) (8). Before 2011, ^{137}Cs levels off Japan were ∼1–2 Bq·m^{-3} (9) (1 Bq = 1 disintegration per second).

Results and Discussion

In June 2011, surface-water concentrations of ^{134}Cs were highly elevated in near-shore areas, although not necessarily at stations closest to the Fukushima Dai-ichi NPPs (Fig. 1*A* and Table S1). The highest ^{134}Cs activities, 3,900 Bq·m^{-3}, were associated with a semipermanent eddy, seen here in the surface drifter data centered on 37°N 142.5°E (Fig. 2*B*). Activities up to 325 Bq·m^{-3} were found more than 600 km from the NPPs (Table S1). The ^{134}Cs:^{137}Cs ratio was close to 1 in all samples clearly indicating a Fukushima NPP source (5) (calculated end-member ratio of 0.975; Fig. S2); the exception to this are the southernmost stations, where we see ^{137}Cs at preexisting levels of 1–2 Bq·m^{-3} and no detectable ^{134}Cs. In our samples, < 0.1% of total Cs was caught on a 1-μm filter, consistent with its more soluble nature (*SI Methods*).

It is evident that the Kuroshio Current forms a southern boundary for the transport of these Fukushima-derived radionuclides (at least in the western North Pacific), because samples at this boundary or to the south had ^{134}Cs activities <3 Bq·m^{-3} (our detection limit for ^{134}Cs was 1.5 Bq·m^{-3}). This finding is supported by the tracks of our surface drifters, most of which traveled east along the Kuroshio or northeast (Fig. 2*A*, blue and red lines). However, several drifters moved westward toward the coast, with one drifter transiting along the coast to the south of our study area (Fig. 2*B*, magenta lines), suggesting a potential pathway for contaminated water. Although we have no Cs

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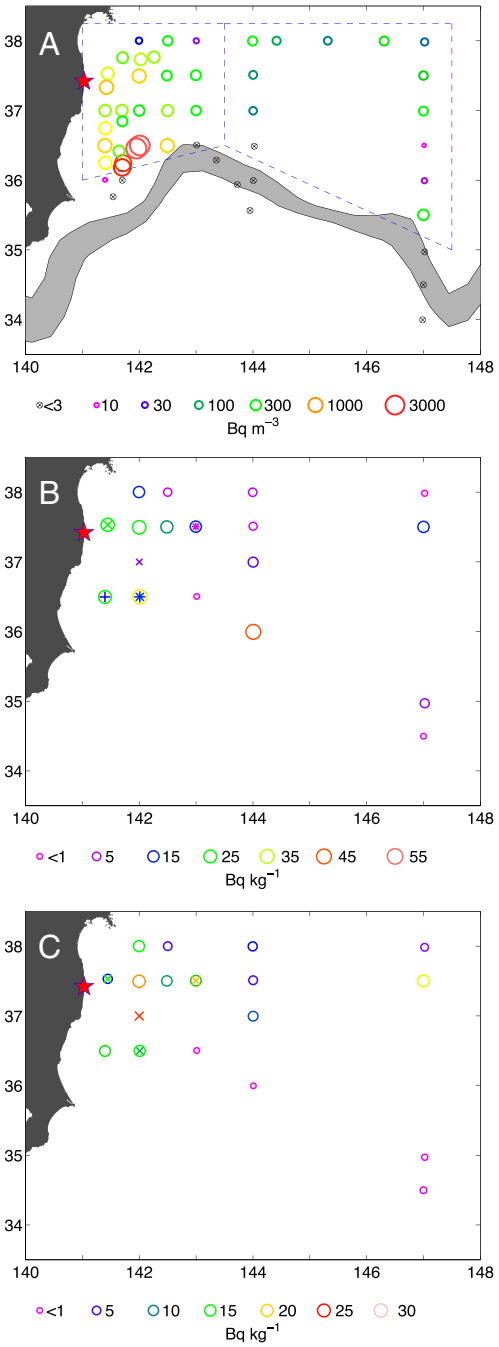

Fig. 1. (*A*) Concentrations of ^{134}Cs in surface water (Bq·m^{-3}), (*B*) ^{134}Cs in biological samples (Bq·kg^{-1} dry weight), and (*C*) ^{110m}Ag in biological samples (Bq·kg^{-1} dry weight). Biological samples were separated into mixed zooplankton, crustaceans, and fish as indicated by legend (*SI Discussion*). Red star is location of Fukushima NPPs. Gray shaded area in *A* shows approximate position of Kuroshio Current during the cruise (Fig. S1). Dashed lines in *A* are areas used to calculate near-shore and offshore Cs inventories.

measurements this close to shore, this is consistent as well with the coastal model of Tsumune et al. (4).

^{137}Cs and ^{134}Cs in biota ranged from below detection to 56 Bq·kBq·kgg^{-1} dry weight (Fig. 1*B* and Table 1), depending primarily on the sampling location but also on the biological composition of the sample. As in the water, ^{134}Cs:^{137}Cs ratios were essentially 1. The only other artificial gamma-emitting radionuclide detected consistently in biota was ^{110m}Ag (Fig. 1*C*),

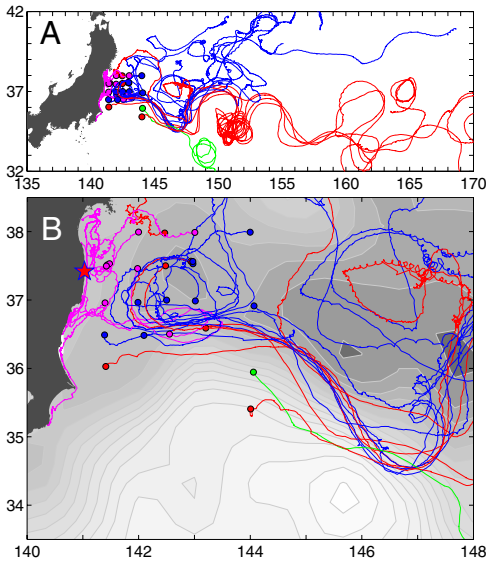

Fig. 2. (*A*) Tracks of surface drifters released at the sampling stations, from time of release through October 26, 2011. Trajectories are color coded: blue drifters traveled north of the Kuroshio, red were advected east in the Kuroshio, green went south of the Kuroshio, and magenta stayed close to the coast. (*B*) Expanded view of coastal region showing more clearly the near-shore eddies and coastal transport.

which was detected in zooplankton samples with concentrations up to 23.6 Bq·kg^{-1} dry weight, but was undetectable in all three fish samples (Table 1). Overall, Ag is known to be highly particle-reactive in seawater and greatly concentrates in both phytoplankton, at the base of the food chain, and zooplankton (10), consistent with the data in Table 1. The ^{110m}Ag concentrations in plankton samples north of the Kuroshio were lowest in samples more distant from Japan. The source of the ^{110m}Ag measured in zooplankton is unclear; no direct releases to the coastal ocean have been reported, but soils collected near the power plants did show trace levels of ^{110m}Ag (11). In zooplankton, ^{134}Cs:^{110m}Ag ratios ranged up to 4.7, with most values 0.8–2. Gelatinous zooplankton, collected at one station, had about half the radioactivity of crustacean zooplankton at that same station for either Cs isotope or ^{110m}Ag.

Concentration factors—essentially the degree of radionuclide enrichment in biota relative to ambient water—are used to evaluate the radiological risks associated with seafood consumption and were determined by dividing the radionuclide concentration in biota by the dissolved concentration in surface water. Median values were 44 for ^{137}Cs and 36 for ^{134}Cs, comparable to the recommended International Atomic Energy Agency (IAEA) value of 40 for zooplankton (10). Concentration factors for ^{110m}Ag were not calculated because dissolved concentrations could not be measured using our seawater method, which was specific to Cs isotopes only.

The median concentration of ^{137}Cs in micronektonic fish (secondary consumers) was less than that in zooplankton (primary and secondary consumers) and about 150-fold below the Japanese legal limit for fish of 500 Bq·kg^{-1} wet weight. Compared with oceans that were most heavily impacted by Chernobyl, our ^{137}Cs values in fish are equivalent to those found in the Black Sea, but less than in the Baltic Sea, where fish concentrations varied with locations (12). The combined gamma radioactivity of ^{134}Cs, ^{137}Cs, and ^{110m}Ag in biota ranged from 1% to 54% (typically 10–30%) of total radioactivity attributable to naturally occurring radionuclides, of which ^{40}K contributed >99% of the total (Table 1).

Table 1. Concentrations of naturally occurring ^{40}K and Fukushima-derived radionuclides (^{137}Cs, ^{134}Cs, and ^{110m}Ag) in marine organisms collected off Japan's coast (Bq·kg^{-1} dry weight)

Sampling method	Station	Sample composition		Abundance of dominant species, %	^{137}Cs Bq·kg^{-1}, dry weight	^{134}Cs Bq·kg^{-1}, dry weight	^{137}Cs CF	^{134}Cs CF	^{110m}Ag Bq·kg^{-1}, dry weight	^{40}K Bq·kg^{-1}, dry weight	^{134}Cs:^{137}Cs	^{134}Cs:^{110m}Ag	Anthropogenic: natural radioactivity ratio
Bongo nets, 300 μm	2	Zooplankton mixed	Copepods	50	0.3	bd	134	bd	1.9	217	bd	bd	0.01
	3			70	6.4	6.2	*	bd	1.3	229	1.0	4.7	0.06
	8			80	14.3	15.1	98	105	18.3	321	1.1	0.8	0.07
	9			92	1.2	1.2	16	17	3.3	156	1.0	0.4	0.04
	10			71	4.7	4.6	27	26	6.5	147	1.0	0.7	0.11
	11			61	3.1	3.9	29	36	5.3	126	1.3	0.7	0.10
	12			72	7.2	8.8	77	98	8.7	148	1.2	1.0	0.17
	14			65	56.4	45.5	*	*	0.0	188	0.8	bd	0.54
	17			47	0.9	bd	255	bd	0.5	195	bd	bd	0.01
	19			65	12.8	15.0	53	59	12.0	232	1.2	1.2	0.17
	21			52	17.3	20.7	78	88	11.1	225	1.2	1.9	0.22
	22			79	4.5	4.2	20	19	5.1	129	0.9	0.8	0.11
	23			58	29.1	31.4	34	36	21.5	181	1.1	1.5	0.45
	24			65	12.8	15.5	285	351	14.0	197	1.2	1.1	0.21
	25			51	26.2	29.4	44	49	6.3	272	1.1	4.7	0.23
	27			84	19.9	25.4	27	33	12.4	194	1.3	2.0	0.30
	29			76	34.2	38.0	10	10	14.4	230	1.1	2.6	0.38
				Median	12.8	15.1	44	36	6.5	195			
				Mean	14.8	17.7	79	71	8.4	199			
				SD	14.9	13.7	85	90	6.4	51			
Methot net, 4 mm	25	Scyphomedusan	*Chrysaora* sp.	100	11.1	11.9	19	20	3.4	197	1.1	3.5	0.13
	19	Euphausiids	*Euphausia pacifica*	88	2.3	4.5	9	18	20.6	196	2.0	0.2	0.14
	25			100	23.4	27.2	39	45	15.7	214	1.2	1.7	0.31
	29	Euphausiids	*Nematoscelis difficilis*	86	15.1	16.3	4	4	11.5	182	1.1	1.4	0.24
	32	Fish, shrimps, euphausiids, pyrosomatids mixed	nd	nd	3.1	6.8	9	20	23.6	278	2.2	0.3	0.12
				Median†	9.1	11.6	9	19	18.2	205			
				Mean†	11.0	13.7	16	22	17.9	217			
				SD†	10.1	10.3	16	17	5.3	42			
	19	Fish	*Stenobrachius leucopsarus*	50	4.9	3.3	20	13	bd	144	0.7	bd	0.06
	27	Fish	*Cyclothone pseudopallida, Sigmops gracile*	40, 40	11.4	11.9	16	15	bd	232	1.0	bd	0.10
	29	Fish	*S. gracile*	63	15.8	14.8	4	4	bd	129	0.9	bd	0.24
				Median	11.4	11.9	16	13	bd	144			
				Mean	10.7	10.0	13	11	bd	168			
				SD	5.5	6.0	8	6	bd	55			

Natural radioactivity refers to the sum of gamma radioactivity emitted by ^{40}K (>99% of total natural radioactivity), ^{212}Pb, ^{235}U, ^{211}Bi, and ^{7}Be. Concentration factors (CFs) were calculated using surface ocean concentrations (Table S1). The biological composition of the samples is shown as percent of total abundance. bd, below detection; nd, not determined; sp, species unidentified.

*In this sample, CFs of ^{134}Cs and ^{137}Cs are five orders of magnitude higher due to very low surface ocean Cs, and therefore not included in mean or median CF values.

†Considers nongelatinous zooplankton only.

The surface ocean Cs data leads to several key conclusions regarding the impact of the Fukushima Dai-ichi NPPs on the ocean. First, Fukushima-derived ^{134}Cs and ^{137}Cs were measured at up to 1,000× higher activities than existed previously and were found throughout a 150,000-km^2 area of the Pacific Ocean off Japan. Second, though elevated, substantial dilution had occurred between the discharge channels at the Fukushima Dai-ichi NPPs, where ^{137}Cs activities averaged 33,000 Bq·m^{-3} in June (5) and our closest samples 30 km offshore, which were on average 50× lower (600–800 Bq·m^{-3}). Our Cs activities are thus consistent with the Japanese reporting in June "below detection" for ^{134}Cs and ^{137}Cs 30 km off-shore [Japanese Ministry of Education, Sports, Science and Technology (MEXT)], because their methods had a higher detection limit of 10,000 Bq·m^{-3} (5). As a result of ocean stirring and mixing processes, all ^{137}Cs activities offshore have been reduced to well below the Japanese regulatory limits for the ocean (90,000 Bq·m^{-3}) and are below the levels of the most abundant natural radionuclide in the ocean, ^{40}K (~12,000 Bq·m^{-3}).

We measured ^{134}Cs activities decreasing with depth, and as with the surface data, activities in the subsurface are generally higher near-shore than offshore (Fig. 3). Prior studies show penetration of 1960s fallout radionuclides, including ^{137}Cs, down to 1,000 m in this area (9), but in the short time here since its release, Fukushima-derived Cs does not generally penetrate below ~100–200 m. Over time, we expect to see deeper penetration and lateral mixing for Cs and other soluble Fukushima isotopes, providing a means to study the rates of vertical and horizontal mixing processes in the Pacific.

Cesium profiles are essential to calculate ocean contamination for comparison with land, and to calculate the total Cs inventory in our study area for comparison with the widely varying source estimates. When integrated vs. depth, aerial inventories of ^{137}Cs at different locations in the near-shore area range from 7,000 to 80,000 Bq·m^{-2}, decreasing to 200–14,000 Bq·m^{-2} in the offshore stations (areas denoted in Fig. 1*A*; *Methods*). Preexisting ^{137}Cs inventories in these waters would be ~200–300 Bq·m^{-2} (down to 200 m) (9). It is important to note that our highest values are still 40× lower than the highest estimated aerial burdens on land within 30 km of the NPPs (3 million Bq·m^{-2} ^{137}Cs; ref. 13, http://radioactivity.mext.go.jp/en/1750/2011/08/1750_083014.pdf).

There has been considerable debate about both the total radionuclide releases and the extent of atmospheric fallout vs. direct discharges from the Fukushima NPPs. Total atmospheric releases of ^{137}Cs have been estimated by several Japanese groups and official sources to range from 10 to 13 PBq (1 PBq = 1 × 10^{15} Bq; Table S2), with peak discharges delivered March 15–25, 2011 (1, 2). From the 10-PBq total atmospheric source, Morino et al. (2) suggest that 22% fell on land in Japan and 10% (1 PBq) in their ocean domain (defined out to 145°E), with about 50% advecting farther east over the ocean. A more recent study by Stohl et al. (6) found a similar fraction on land, but suggested much higher total atmospheric releases for ^{137}Cs of 36 PBq (23–50 PBq).

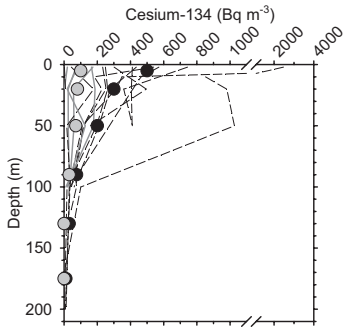

Fig. 3. Profiles of ^{134}Cs (Bq·m^{-3}) vs. depth for offshore (gray solid lines; mean, gray circles) and near-shore stations (black dashed lines; mean, black circles).

Direct discharges were an additional radionuclide source to the ocean, dominated by a leak from Dai-ichi reactor 2 April 1–6, 2011. Similar to atmospheric fallout, there is a considerable range of release estimates, from 3.5 PBq (4) to as high as 22 PBq (Table S2). We measured a total inventory of Fukushima-derived ^{137}Cs of 1.9–2.1 PBq in our study area (*Methods*), which is lower because most of the ^{137}Cs delivered either as fallout in March or direct discharges in April would have been transported out of the study area by June.

To quantify this transport and assess if our data support the higher or lower release estimates, we modeled the spreading of contaminated waters by ocean currents (*Methods* and *SI Discussion*). The model reproduced the measured ^{137}Cs distribution in June, with the largest concentrations associated with a near-shore cyclonic eddy, and intermediate concentrations at stations closest to shore, resulting from later stages of direct discharges, and the southern edge of contaminated waters aligning with the Kuroshio (Fig. S3). The model predicts an inventory of ~0.4 PBq (using the lowest atmospheric and direct discharge source estimates) to ~2.0 PBq (using the highest estimates) in our study area in June (*SI Discussion*). Thus, if we include realistic transport, our measured inventory in June agrees better with what is predicted using the largest release estimates; this is important, but needs to be supported by additional data over larger areas of the North Pacific. However, at present, such data are extremely limited and insufficient to make a basin-wide inventory calculation. A more complex 3D ocean model would also be helpful to quantify transport processes and predict long-term concentration trends expected throughout the Pacific.

In terms of potential biological impacts, radiation doses in marine organisms are generally dominated by the naturally occurring radionuclides ^{210}Po (an alpha emitter) and ^{40}K, even when organisms are exposed to anthropogenic radioactivity discharged to coastal waters (14). To be comparable just to doses from ^{210}Po, ^{137}Cs levels in fish would need to range from 300 to 12,000 Bq·kg^{-1} dry weight, some 1–3 orders of magnitude higher than what we observed ≥30 km off Japan. Thus, radiation risks of these isotopes to marine organisms and human consumers of seafood are well below those from natural radionuclides (*SI Discussion*). Further, we can calculate an external dose to humans if immersed/swimming in these waters of <0.01 μSv·d^{-1} at ^{134}Cs and ^{137}Cs levels of 1,000 Bq·m^{-3}, which is <0.3% of the average Japanese dose of about 4 μSv·d^{-1} from all radioactivity sources. Finally, these levels are several orders of magnitude lower than those used in one study that assumed exposure to the most heavily impacted water discharged from the Fukushima NPPs to predict marked reproductive effects and possible mortality in marine biota (15).

Several other gamma-emitting fission products (e.g., ^{106}Ru, ^{103}Ru, ^{144}Ce, and ^{141}Ce) were detected after Chernobyl in biota and sinking particles (16), but were not seen here in biota (or on filters), or by others in soils around the Fukushima NPP (11). These refractory isotopes were prevalent from Chernobyl due to the massive explosion and release of core materials in addition to radioactive gases and volatiles. The Fukushima accident was characterized by core overheating that led to the venting of radioactive gases, hydrogen explosions, and fires associated with spent fuel rods; this resulted in the preferential release of more volatile radionuclides, such as Cs, and gases to the atmosphere. However, the salt and fresh water used to cool the Fukushima NPPs and acidic conditions in the core provide possible pathways to mobilize refractory radionuclides from the core that may have subsequently been discharged to the ocean but have yet to be assessed. Ultimately, though the radionuclide levels of ^{137}Cs and

^{134}Cs offshore are currently low with respect to human health effects, any assessment of radiation dose should also consider long-term exposure if the NPP remains a continued source of radionuclides (5) and if, as has been reported, coastal sediments are contaminated with multiple radionuclides.

Methods

Details of sampling, analytical methods, transport model, and calculations of dose are provided in *SI Discussion*, with a brief summary below.

Method for ^{134}Cs and ^{137}Cs in Seawater. Seawater samples were extracted onto an ion exchange resin made of the organic polymer polyacrylonitrile (PAN) and ammonium molybdophosphate (AMP) (17). Resin was gamma counted for ^{134}Cs and ^{137}Cs isotopes using high-purity germanium well detectors in the laboratory. Resin extraction recoveries were determined from initial and final sample aliquots containing stable Cs added as a yield monitor. Calibration standards were run using archived water from the Sargasso Sea and an International Atomic Energy Agency Irish Sea water reference report (IAEA-443) (18). The concentration of Cs isotopes in water is reported in units of becquerels per unit volume (Bq·m^{-3}). All activities are decay corrected to April 6, 2011, the date of the maximum direct radioactivity discharge into the ocean (5).

Biota Sampling and Analysis. We obtained samples of the zooplankton and nekton community by double-oblique tows of Bongo nets (0.6 m diameter, 0.3 mm mesh) and Methot trawls (2.5 × 2.5 m frame, 4 mm mesh). Aliquots (1–10% of total volume) of the fresh samples were preserved with formalin and sorted into higher taxa, enumerated, and converted to abundance (details in *SI Methods*). Remaining samples were immediately frozen and later freeze-dried before radioanalysis by gamma spectrometry using a low-energy germanium detector. Radionuclide calibrations were made against a certified IAEA fish standard (19). Radioactivity was decay corrected to April 6, 2011, similar to seawater. Radionuclide concentration factors were based on surface-water concentrations because these animals were exposed to higher ^{134}Cs and ^{137}Cs concentrations while feeding at night in the surface, and because these animals were mostly caught during night tows. Dose estimates are discussed further in *SI Discussion*.

Drifter Methods/Data Source. A total of 24 surface drifters were deployed at 20 of the stations. The drifters, manufactured by Pacific Gyre, are drogued at 15 m, and also measure sea-surface temperature. The locations of the drifters were relayed numerous times per day using the Argos satellite tracking system and archived at the Global Drifter Program Drifter Data Assembly Center (http://www.aoml.noaa.gov/phod/dac/dacdata.php).

Transport Model. The velocity fields used to simulate the spreading of contaminated waters were derived from satellite altimetry data and National Centers for Environmental Prediction/National Center for Atmospheric Research reanalysis products. The Ekman velocities due to winds were added to the geostrophic velocities. To estimate the spreading of contaminated waters, we used the Lagrangian advection scheme with a variable-step Runge–Kutta 4 (20) integration and a bilinear interpolation in time and space between grid points. Further details of the transport model are provided in *SI Methods*. Also, as discussed in the text and further in *SI Methods*, the source function to the model is either atmospheric deposition to the coast north of the NPP peaking March 17–20, 2011 (red rectangle in Fig. S3 *A–D*), or direct ocean discharges using at point source at the Fukushima NPP and the measured time-series of input reported in Buesseler et al. (5) that peaked on April 6, 2011 (Fig. S3 *E–H*).

Inventory Calculations. Ocean inventories of ^{137}Cs were determined by simple trapezoidal integration of the ^{137}Cs vs. depth profiles, and these averaged 26,000 Bq·m^{-2} and 6,200 Bq·m^{-2} for near-shore and offshore areas in Fig. 1*A*, respectively. When multiplied by the area of each, we obtained a total inventory of 1.25 PBq for the near-shore area (50,000 km^2) and 0.62 PBq for the offshore area (100,000 km^2), or a total of 1.9 PBq in our study area. Alternatively, with the exception of two stations, there is a significant linear relationship between total ^{137}Cs inventory and the surface activities [surface ^{137}Cs (Bq·m^{-3}) × 42 = ^{137}Cs inventory (Bq·m^{-2}); R^2 = 0.63], and if we use this relationship and the better aerial coverage provided by the larger set of locations where we have surface ^{137}Cs data only, we come up with a similar total inventory of 2.1 PBq. Thus, for this article, we use 2.0 PBq as the measured total inventory of ^{137}Cs in our study area.

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1. Chino M, et al. (2011) Preliminary estimation of release amounts of 131I and 137Cs accidentally discharged from the Fukushima Daiichi nuclear power plant into the atmosphere. *J Nucl Sci Technol* 48:1129–1134.
2. Morino Y, Ohara T, Nishizawa M (2011) Atmospheric behavior, deposition, and budget of radioactive materials from the Fukushima Daiichi nuclear power plant in March 2011. *Geophys Res Lett* 38:L00G11.
3. Yasunari TJ, et al. (2011) Cesium-137 deposition and contamination of Japanese soils due to the Fukushima nuclear accident. *Proc Natl Acad Sci USA* 108:19447–19448.
4. Tsumune D, Tsubono T, Aoyama M, Hirose K (2011) Distribution of oceanic (137)Cs from the Fukushima Dai-ichi nuclear power plant simulated numerically by a regional ocean model. *J Environ Radioact*, 10.1016/j.jenvrad.2011.10.007.
5. Buesseler KO, Aoyama M, Fukasawa M (2011) Impacts of the Fukushima nuclear power plants on marine radioactivity. *Environ Sci Technol* 45:9931–9935.
6. Stohl A, et al. (2011) Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: Determination of the source term, atmospheric dispersion, and deposition. *Atmos Chem Phys Discuss* 11:28319–28394.
7. Bailly du Bois P, et al. (2011) Estimation of marine source-term following Fukushima Dai-ichi accident. *J Environ Radioact*, 10.1016/j.jenvrad.2011.11.015.
8. Aarkrog A (2003) Input of anthropogenic radionuclides into the World Ocean. *Deep Sea Research Part II: Topical Studies in Oceanography* 50(17–21):2597–2606.
9. Aoyama M, Hirose K (2004) Artificial radionuclides database in the Pacific Ocean: HAM database. *ScientificWorldJournal* 4:200–215.
10. International Atomic Energy Agency (IAEA) (2004) Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment (IAEA, Vienna), Technical Report Series No. 422.
11. Tagami K, et al. (2011) Specific activity and activity ratios of radionuclides in soil collected about 20 km from the Fukushima Daiichi nuclear power plant: Radionuclide release to the south and southwest. *Sci Total Environ* 409:4885–4888.
12. International Atomic Energy Agency (IAEA) (1995) Sources of Radioactivity in the Marine Environment and Their Relative Contributions to Dose Assessment from Marine Radioactivity (MARDOS) (IAEA, Vienna), TECDOC 838.
13. Japanese Ministry of Education, Sports, Science and Technology, MEXT (2011) Available at http://radioactivity.mext.go.jp/en/1750/2011/08/1750_083014.pdf. Accessed March 2012.
14. Aarkrog A, et al. (1997) A comparison of doses from 137Cs and 210Po in marine food: A major international study. *J Environ Radioact* 34:69–90.
15. Garnier-Laplace J, Beaugelin-Seiller K, Hinton TG (2011) Fukushima wildlife dose reconstruction signals ecological consequences. *Environ Sci Technol* 45:5077–5078.
16. Buesseler KO, et al. (1987) Chernobyl radionuclides in a Black Sea sediment trap. *Nature* 329:825–828.
17. Sebesta F, Stefula V (1990) Composite ion exchanger with ammonium molybdophosphate and its properties. *J Radioanal Nucl Chem* 140:15–21.
18. Pham M, et al. (2011) A certified reference material for radionuclides in the water sample from Irish Sea (IAEA-443). *J Radioanal Nucl Chem* 288:603–611.
19. Pham MK, et al.; International Atomic Energy Agency (2006) Certified reference material for radionuclides in fish flesh sample IAEA-414 (mixed fish from the Irish Sea and North Sea). *Appl Radiat Isot* 64:1253–1259.
20. Ralph EA, Niiler PP (1999) Wind-driven currents in the tropical Pacific. *J Phys Oceanogr* 29:2121–2129.